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IN THE UNITED STATES PATENT & TRADEMARK OFFICE

IN RE APPLICATION OF :  
HIROYUKI KURIMURA, ET AL. : EXAMINER: MULLIS, JEFFREY C.  
SERIAL NO: 10/549,572 :  
FILED: SEPTEMBER 19, 2005 : GROUP ART UNIT: 1796  
FOR: BLOCK COPOLYMER MIXTURE :  
CONTAINING STAR-BRANCHED  
BLOCK COPOLYMER

DECLARATION UNDER 37 C.F.R. §1.132

COMMISSIONER FOR PATENTS  
ALEXANDRIA, VIRGINIA 22313

SIR:

I, Jun Yoshida state:

1. That I am a graduate of the University of Tokyo and received my Masters degree in the year 1999.
2. That I have been employed by Denki Kagaku Koray Kabushiki Kalsha for 9 years as a Researcher in the field of Polymers.
3. That I have read and understood Moczygemba et al., US 5,705,569 (Moczygemba I).
4. The following experiments were performed by me or under my supervision.
5. The following experiments demonstrate that the claimed block copolymer mixture is advantageous in balance between transparency and impact resistance to that of Moczygemba I.

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#### 6. Experimental Data

The experiments were conducted by the polymerization method disclosed as the third embodiment in Table 19 of Moczygemba I and the polymerization method disclosed in Example 1 of the present invention, respectively.

#### 7. Experiment 1 (TABLE 19 in Moczygemba I)

(1) Into a reactor, 467 kg of a polymerization solvent was charged and maintained at 30°C.

(2) As a polymerization catalyst solution, 850 ml of an n-butyllithium 10 mass% cyclohexane solution was added thereto, followed by adding 80.0 kg of a styrene monomer to carry out anionic polymerization of a styrene monomer. Hereinafter, as all of polymerization catalyst solutions in an experimental data, an n-butyllithium 10 mass% cyclohexane solution was employed.

(3) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 910 ml of a polymerization catalyst solution was added and further 40.0 kg of a styrene monomer was added thereto to carry out a reaction.

(4) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 2670 ml of a polymerization catalyst solution was added and further 20.0 kg of a styrene monomer was added thereto to carry out a reaction.

(5) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 30.0 kg of a styrene monomer was added thereto to carry out a reaction.

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(6) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 30.0 kg of butadiene was added thereto to carry out a reaction:

(7) After butadiene gas was completely consumed, while the internal temperature of the reaction system was maintained at 75°C, a solution obtained by dissolving 860 g of a coupling agent "ADEKASIZER O-130PTM" (manufactured by ASAHI DENKA CO., LTD) made of epoxidized soybean oil in 5L of cyclohexane, was added thereto to carry out a coupling reaction for 10 minutes.

(8) All polymerization active terminals were deactivated by water.

(9) After a preliminary concentration of the polymerization solvent, an evaporation treatment was carried out by a vented extruder to obtain pellets.

8. *Experiment 2 (Example 1 of the present invention)*

(1) Into a reactor, 467 kg of a polymerization solvent was charged and maintained at 30°C.

(2) 710 ml of a polymerization catalyst solution was added thereto, followed by adding 69.0 kg of a styrene monomer to carry out anionic polymerization of a styrene monomer.

(3) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 1700 ml of a polymerization catalyst solution was added and further 23.0 kg of a styrene monomer was added thereto to carry out a reaction.

(4) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 3560 ml of a polymerization catalyst solution was added and further 52.0 kg of a styrene monomer was added thereto to carry out a reaction.

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(5) After the styrene monomer was completely consumed, while the internal temperature of the reaction system was maintained at 50°C, 56.0 kg of butadiene was added thereto to carry out a reaction.

(6) After butadiene gas was completely consumed, while the internal temperature of the reaction system was maintained at 75°C, a solution obtained by dissolving 718 g of a coupling agent "ADEKASIZER O-130PTM" (manufactured by ASAHI DENKA CO., LTD) made of epoxidized soybean oil in 5L of cyclohexane, was added thereto to carry out a coupling reaction for 10 minutes.

(7) All polymerization active terminals were deactivated by water.

(8) After a preliminary concentration of the polymerization solvent, an evaporation treatment was carried out by a vented extruder to obtain pellets.

9. The block copolymer mixture obtained in each of Experiments 1 and 2 was blended with a general-purpose polystyrene (manufactured by Toyo Styrene Co., Ltd.: G200C) at a weight ratio of the block copolymer mixture/general-purpose polystyrene of 6 to 4, followed by supplying to a 40 mm single screw extruder, and a molten strand was withdrawn from a die at 200°C, cooled with water and cut by a cutter to obtain resin pellet.

10. Further, molecular weight data in tables 2 to 5, and MFR, Haze, Total luminous transmittance, Total absorbed energy and Charpy impact strength in Table 6 were measured in the same manner as disclosed in the specification of the present application.

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Table 1 Amount of materials changed

|                               | Experiment 1 | Experiment 2 | Disclosed value in Table 19 of Moczygemba I    | Disclosed value in Example 1 of the present invention |
|-------------------------------|--------------|--------------|--|---|
| Solvent cyclohexane (g)       | 467 kg       | 467 kg       | 168.9 phm                                      | 3836 g  |
| First stage catalyst solvent  | 850 ml       | 710 ml       | 0.032 phm                                      | 5.8 ml  |
| First stage styrene           | 80.0 kg      | 69.0 kg      | 40 phm   | 568 g   |
| Second stage catalyst solvent | 910 ml       | 1700 ml      | 0.0034 phm                                     | 14 ml   |
| Second stage styrene          | 40.0 kg      | 23.0 kg      | 20 phm   | 188 g   |
| Third stage catalyst solvent  | 2670 ml      | 3560 ml      | 0.100 phm                                      | 29.3 g  |
| Third stage styrene           | 20.0 kg      | 52.0 kg      | 10 phm   | 428 g   |
| Fourth stage styrene          | 30.0 kg      | -            | 15 phm   | -   |
| Butadiene                     | 30.0 kg      | 56.0 kg      | 15 phm (0.5 phm is considered to be incorrect) | 460 g   |
| Epoxidized soybean oil        | 860 g        | 718 g        | 0.42 phm                                       | 5.9 g   |

Table 2 Analyzed values of polymer content formed by the end of fourth stage styrene polymerization

|                                       | Experiment 1 | Experiment 2 | Disclosed value in Example 1 of the present invention |
|---------------------------------------|--------------|--------------|---|
| Peak top molecule weight M1           | 151000       | 140000       | 143000  |
| Peak top molecular weight M2          | 34400        | 17900        | 20000   |
| Peak top molecular weight M3          | 9600         | 7400         | 8900  |
| Molar ratio of component S1           | 16.1         | 9.6          | 9.8   |
| Molar ratio of component S2           | 24.0         | 33.3         | 30.9  |
| Molar ratio of component S3           | 59.9         | 57.1         | 59.3  |
| M1/M3                                 | 15.73        | 18.92        | 16.07   |
| M2/M3                                 | 3.58         | 2.42         | 2.25  |
| Molecular weight distribution (Mw/Mn) | 3.02         | 4.41         | 3.76  |

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Table 3 Analyzed values of polymer content formed by the end of butadiene polymerization

|   | Experiment 1 | Experiment 2 | Disclosed value in Example 1 of the present invention |
|---|--------------|--------------|---|
| Peak top molecular weight M4  | 161000       | 155000       | 167000  |
| Peak top molecular weight M5  | 46400        | 35300        | 41000   |
| Peak top molecular weight M6  | 22000        | 24100        | 29000   |
| M4/M6   | 7.32         | 6.43         | 5.76  |
| M5/M6   | 2.11         | 1.46         | 1.41  |
| Number average molecular weight of polymer block comprising conjugated diene as monomer units | 6800         | 10300        | 11000   |

Table 4 Analyzed values of block copolymer mixture containing a branched block copolymer

|   | Experiment 1 | Experiment 2 | Disclosed value in Example 1 of the present invention |
|---|--------------|--------------|---|
| Weight average molecular weight by GPC  | 144000       | 144000       | 137600  |
| Number average molecular weight by GPC  | 87400        | 86800        | 82100   |
| Molecular weight distribution by GPC  | 1.65         | 1.65         | 1.68  |
| Peak top molecular weight of maximum peak by GPC  | 185000       | 202000       | 200000  |
| Molecular weight distribution ( $M_w/M_n$ ) of peak at which peak top molecular weight becomes minimum among peaks (a) at which the peak top molecular weight is from 20,000 to 50,000 and (b) which form a proportion of the area of from 3 to 15% | 1.028        | 1.033        | 1.008   |
| Proportion (%) of the area of peak at which the peak top molecular weight becomes maximum among peaks at which the peak top molecular weight is from 200,000 to 380,000   | 7.1          | 43.0         | 5.2   |

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Table 5 Results of evaluation of physical properties of block copolymer mixture containing a branched block copolymer

|                                  | Experiment 1 | Experiment 2 | Disclosed value in Example 1 of the present invention |
|----------------------------------|--------------|--------------|---|
| PBD amount (%)                   | 15           | 28           | 28  |
| MFR (g/10 min)                   | 10.0         | 13.1         | 15.5  |
| Total luminous transmittance (%) | 90.4         | 89.2         | 89  |
| Haze (%)                         | 4.8          | 8.2          | 2.0   |

Table 6 Blend of block copolymer mixture containing a branched block copolymer and general-purpose polystyrene

|                                  | Experiment 1 | Experiment 2 |
|----------------------------------|--------------|--------------|
| MFR (g/10 min)                   | 8.4          | 9.1          |
| Haze (%)                         | 3.9          | 11.1         |
| Total luminous transmittance (%) | 89.2         | 74.6         |
| Total absorbed energy (J)        | 0.05         | 0.15         |
| Charpy impact strength (kJ/m)    | 1.4          | 1.3          |

11. As shown in Table 6, the total absorbed energy is 0.05 J in the experiment of Moczygemba I, while the total absorbed energy is 0.015 J in the experiment of the present specification. A poor impact resistant polymer of Moczygemba I is insufficient for an application for extrusion molding for e.g. sheets or films.

12. With the process describe in the present specification, the claimed copolymer is obtained in which the balance between the transparency and the impact resistance is advantageous. The claimed block copolymer mixture cannot be readily obtained based on the disclosure of Moczygemba I.

13. The undersigned declare further that all statements made herein are of his own knowledge are true and that all statements made on information are believed to be true. Further that these statements were made with the knowledge that willful false statements and

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the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of this application or any patent issuing thereon.

*Jun Yoshida*  
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Signature

*Nov. 13 , 2008*  
\_\_\_\_\_  
Date